# Spin quantum beats of hot trion in quantum dots

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**Abstract.** Spins of resident holes in charged quantum dots (QD's) act as local magnets inducing Zeeman splitting of excitons trapped into dots. This has been evidenced by observation of quantum beats in linearly polarized time-resolved photoluminescence of a biased array of self-assembled InP QD's. A theory is developed that considers the exchange interaction within the positively charged exciton (trion) states formed in the QD.

Electronic and optical properties of zero-dimensional quantum objects are extremely sensitive to the presence of single charge carriers in the system [1–3]. If a neutral quantum dot (QD) is an analog of an atom, a charged quantum dot is an analog of an ion. Peculiarities of singly charged QD's have become a subject of particular attention during recent couple of years, in connection with the problem of quantum computing [4].

Here we report a new spin-related effect in a charged QD. We have observed spin quantum beats (QB's) in linearly polarized photoluminescence (PL) of an array of InP QD's under linearly polarized excitation, while circular polarization of PL under circular-polarized excitation has shown almost no modulation. Such kind of beats is a signature of splitting between exciton resonance energies in $\sigma^+$ and $\sigma^-$ polarizations. In neutral dots, this would have been possible only in a magnetic fields (Zeeman splitting). In our case, such a Zeeman-like splitting is observed in the absence of the external magnetic field. We attribute this unexpected behavior to a peculiar spin structure of the three-particle complex, or trion, formed in the positively charged QD on the exciton creation.

We have studied heterostructures with a single layer of InP self-assembled QD's embedded between $Ga_{0.5}In_{0.5}P$ barrier layers grown by the gas source molecular beam epitaxy. The average base diameter of the QD's was about 40 nm with a height of about 5 nm. The samples were provided with a semi-transparent Shottky contact on the top surface and with an ohmic contact on the back one. The total thickness of undoped layers was about 0.5 $\mu$m. The sample has been illuminated at normal incidence. The PL was excited quasiresonantly, i.e., within the PL band of the QD's, by 3 ps pulses of a mode-locked Ti:sapphire laser. The PL kinetics was measured with a time resolution of 6 ps using a 0.25 m subtractive dispersion double monochromator and a streak-camera. The measurements were done at 5 K.

We can summarize our experimental results as follows. Under certain conditions, namely, at applied bias around some value $U_{\mathrm{opt}}$ and under linearly polarized excitation, PL kinetics of the QD's exhibits pronounced oscillations both in *co-* and *cross*-polarizations (i.e., in polarizations which are identical or orthogonal to that of the incident light, re-

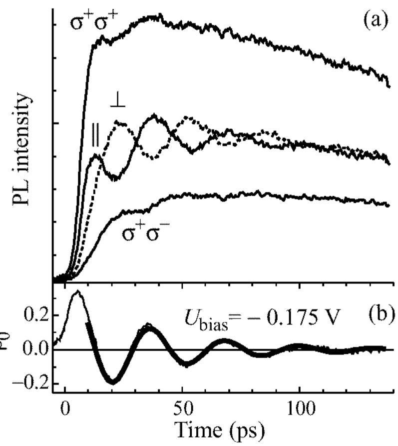


**Fig. 1.** (a) PL kinetics in the circular co- ($\sigma^+\sigma^+$) and cross- ($\sigma^+\sigma^-$) polarizations, and also in the linear co- ($\|$) and cross- ($\perp$) polarizations (indicated against each curve). (b) Degree of linear polarization $\rho_l$ (noisy curve) and the fit (thick gray curve) by equation described in the text.

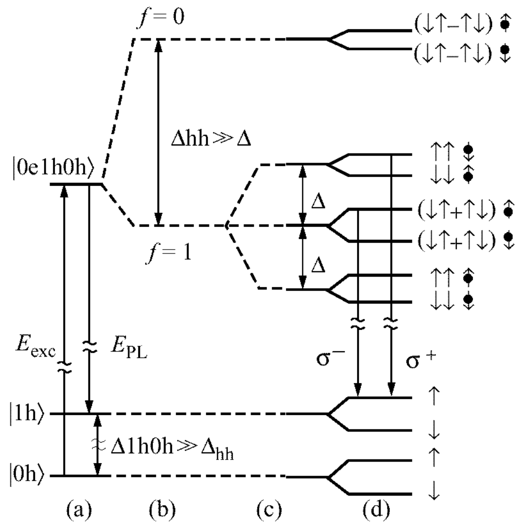


**Fig. 2.** Energy structure of the hot trion. Small arrows ↑ and ⧫ indicates the hole and electron spins, respectively. See further details in the text.

spectively). These oscillations have opposite phases in co- and cross-polarizations thus giving a clear picture of the QB's. When exciting with a circularly polarized light ($\sigma^+$) and detecting either in $\sigma^+$ or $\sigma^-$ circular polarizations, we do not observe any beats. The examples of the PL kinetics are shown in Fig. 1. The degree of linear polarization of the PL, $\rho_l = (I_\| - I_\perp)/(I_\| + I_\perp)$, where $I_\|$ and $I_\perp$ are, respectively, the PL intensities in the co- and cross-polarizations, can be well fitted by the equation $\rho_l(t) = \rho_0 \cos(\omega t)\exp(-t/\tau)$ as shown in Fig. 1(b). The period of the QB's $T = 2\pi/\omega$ is of 30–35 ps, which corresponds to a Zeeman splitting of about 120 $\mu$eV. The decay time of the QB's $\tau \approx 30$ ps which coincides, within the experimental error, with the PL rise time at negative bias $U < U_{\text{opt}}$.

The quantum beats are observed at a Stokes shift $\Delta E < 30$ meV. The beat amplitude is the greatest at $\Delta E = 15$ meV, being strongly dependent on the applied voltage $U$. For instance, at a small deviation of $U - U_{\text{opt}} = \pm 0.15$ V, the beat amplitude drops by a factor of 2. A shift of the excitation energy from short-wavelength to long-wavelength edge of the PL band of the QD's gives rise to insignificant increase of the beat amplitude and to moderate increase of the beat period (from 30 to 50 ps).

To describe the quantum beats, we will consider the following model. Quantum dots contain excess (resident) holes. Their number is controlled by the applied voltage. At $U = U_{\text{opt}}$, one quantum dot contains, in average, exactly one resident hole in its ground state $|0h\rangle$, see Fig. 2(a). Optical excitation created an electron-hole pair in the quantum dot. Note that the electron, in this case, is created in its ground state $|0e\rangle$, and the hole in the excited state $|1h\rangle$. As a result, the quantum dot passes into the state $|0e1h0h\rangle$, which is a hot trion state. The luminescence is caused by recombination of the electron with the resident hole, until the hot hole relaxes into the ground state.

Until the holes are on different energy levels, they may have parallel or antiparallel spins. The spin structure of the hot trion is governed by exchange interactions of the three particles. The holes, being identical particles, interact with each other much stronger than each of them with the electron. The exchange interaction of holes can be recast in the form similar to that of electrons by adopting the pseudospin formalism [5]:

$$\hat{H}_{hh}^{ex} = \Delta_{hh}\left(\vec{j}_1 \cdot \vec{j}_2\right), \tag{1}$$

where the pseudospin $j = 1/2$ has the basic states $\langle 1/2, -1/2|$ and $\langle 1/2, +1/2|$. This interaction gives rise to the singlet-triplet splitting in two levels with the values $f = 0$ and $f = 1$ of the total pseudospin of the two holes ($\vec{f} = \vec{j}_1 + \vec{j}_2$) as shown in Fig. 2(b). The value of the singlet-triplet splitting $\Delta_{hh}$ has been estimated as 3.5 meV for similar QD's by a comparison of model calculations with the experimental single-QD PL spectra [3].

The exchange interaction between an electron and a hole is known to form two spin doublets. The splitting between these doublets, $\Delta_0$, is of the order of 0.1 meV [6, 7]. Since $\Delta_{hh} \gg \Delta_0$, one can consider the exchange interaction of two holes and one electron as interaction of the electron spin $\vec{s}$ with total pseudospin of the two holes $\vec{f}$:

$$\hat{H}_{ehh}^{ex} = 2\tilde{\Delta}_0 f_z s_z + \tilde{\Delta}_1(f_x s_x - f_y s_y) + \tilde{\Delta}_2(f_x s_x + f_y s_y). \tag{2}$$

In what follows, we neglect the two last terms in Eq. (2), since they are insignificant for QB's in linear polarizations. The corresponding energy spectrum of the hot trion comprises four Kramers doublets as shown in Fig. 2(c). Since the energy separation between the $f = 0$ and $f = 1$ level groups is greater than the exciting pulse bandwidth, we can consider them separately. The $f = 0$ doublet can evidently yield no beats. For this reason, the analysis below will be concentrated on the $f = 1$ group of levels. In this group, there are the following doublets (in the $|f_z\rangle|s_z\rangle$ notation): $|\pm 1\rangle|\pm 1/2\rangle$ (radiative), $|0\rangle|\pm 1/2\rangle$ (radiative), and $|\pm 1\rangle|\mp 1/2\rangle$ (non-radiative), see Fig. 2(d). The splitting between these doublets is equal to $\tilde{\Delta}_0$.

Circularly polarized light creates, in the pseudospin notation $|j_z\rangle|s_z\rangle$, the following electron-hole pairs:

$$\sigma^- \longrightarrow |+1/2\rangle|+1/2\rangle \qquad \sigma^+ \longrightarrow |-1/2\rangle|-1/2\rangle \tag{3}$$

Depending on the initial spin state of the resident hole, this results in excitation of different trion states. If the resident hole has $j_z = +1/2$,

$$\sigma^- \longrightarrow |+1\rangle|+1/2\rangle \exp(it\tilde{\Delta}_0/\hbar) \qquad \sigma^+ \longrightarrow |0\rangle|+1/2\rangle \tag{4}$$

If $j_z = -1/2$,

$$\sigma^+ \longrightarrow |-1\rangle|-1/2\rangle \exp(it\tilde{\Delta}_0/\hbar) \qquad \sigma^- \longrightarrow |0\rangle|-1/2\rangle \tag{5}$$

Here $t$ is the time interval after excitation; the energy level of $f_z = 0$ doublet is chosen for zero energy. Radiative transitions from these states give rise to QB's in the PL of QD's if both of these states are excited coherently by the linearly polarized light. Using Eqs. (4) and (5), one can easily derive expressions for the degree of linear polarization of the PL under excitation by linearly polarized light $1/\sqrt{2}\left(\sigma^+ + \sigma^- \exp(2i\varphi)\right)$, where the angle $\varphi$ determines the polarization plane). To do this, one should average intensities of the polarized light components over the spin states of the resident holes in the ensemble of

QD's. Assuming that spins of resident holes are not polarized, we come to the following expression for the degree of linear polarization of PL under linearly polarized excitation:

$$\rho_l(t) \approx 4/5 \cdot \cos\left((\tilde{\Delta}_0/\hbar) \cdot t\right). \tag{6}$$

Note that Eq. (6) is obtained assuming resonant excitation of the $f = 1$ sextet. However, due to inhomogeneous broadening in the QD ensemble, other levels (for instance, the $f = 0$ doublet), bearing no memory of linear polarization, can also be excited in a fraction of dots. This should result in lower value of the PL polarization than it is predicted by Eq. (6). Actually, the beat amplitude of around 0.2 is observed.

The photogenerated hole relaxes to the ground state for around 30 ps. Spins of the two holes, in this case, can be only antiparallel, with their total spin being equal to zero. This is why the fine energy structure of the cold trion is trivial and consists of an electron spin doublet. In this case, no beats can be observed in the absence of magnetic field. For this reason, in the framework of the above model, the beat decay rate coincides with the rate of the hot hole relaxation to its ground state. The luminescence decay time is evidently much larger and lies in the range of several hundreds of ps.

In conclusion, QB's in the PL kinetics of biased heterostructures with InP QD's are studied. The QB's arise due to coherent excitation of spin components of the radiative level. A Zeeman-like spin splitting is shown to be caused by presence of a resident hole inside the QD. The applied electric field controls the charging/discharging of the QD's. A model of the energy structure of hot trion is developed, which explains the main features of the observed QB's.

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